Inelastic neutron scattering around a valence crossover region in $Ce(Cu_{1-x}Ni_x)_2Si_2$

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A heavy-electron compound $CeCu_2Si_2$ is one of the most prominent examples to study a valence crossover phenomenon, which can be induced by applying an accessible pressure or partially substituting Cu with Ni. The valence crossover in $Ce(Cu_{1-x}Ni_x)_2Si_2$ is evidenced by the drastic increase in the Kondo temperature [1, 2]. Indeed, we found that the Kondo temperature shows a step-like increase around $x_c = 0.1$; here the electronic specific heat coefficient, Pauli paramagnetic susceptibility, and T^2-coefficient of the electrical resistivity show an abrupt decrease, indicating the decrease in the electronic density of states. In our evaluation, the Kondo temperature above $x_c \sim 0.1$ is one order larger than that of a lower Ni concentration sample ($x < x_c$). This means the ground state changes from a nearly localized (heavy fermion) to an intermediate valence state. To obtain further insights into this crossover phenomenon, this experiment aims to observe inelastic neutron scattering signals beyond the crossover region.

Polycrystalline samples of $Ce(Cu_{0.8}Ni_{0.2})_2Si_2$ were prepared by arc-melting and annealed at 1373 K in one week. The X-ray diffraction experiments were conducted to check the crystal structure and phase homogeneity. The neutron scattering experiment was carried out at 6G-TOPAN, JRR-3. The incident neutrons were monochromatized with vertically focused PG(002) crystals. Collimation of blank-30'-30'-60' was used. To reduce the higher harmonics, a PG filter was used just before the analyzer. The scattered neutron energy was fixed at 13.4 meV and analyzed with vertically focused PG(002) crystals. Samples were sealed into a standard aluminum cell with a helium exchange gas. A closed-cycle refrigerator was used to cool samples. The inelastic neutron scattering spectra were measured at 10 and 293 K.

Figure 1 shows the inelastic neutron scattering spectra of $Ce(Cu_{0.8}Ni_{0.2})_2Si_2$; the blue and red points indicate the results at 10 and 293 K, respectively. Here the background from the empty cell was subtracted. No well-defined inelastic signal was observed in the measured energy region (0-16 meV), indicating that the system undergoes a nonmagnetic ground state. High-energy signals are likely due to phonon excitations. These results indicate that the spectral weight of the magnetic excitations may move to high energies in the $x > x_c$ samples owing to the drastic increase in the coherence temperature of the 4f-hybridization band. Further neutron investigation of the effect of Ni substitution in $Ce(Cu_{1-x}Ni_x)_2Si_2$ will shed light on the change in the electronic state due to valence crossover.

[1] Y. Ikeda, S. Araki, T.C. Kobayashi, Y. Shimizu, T. Yanagisawa, and H. Amitsuka: J. Phys. Soc. Jpn. **81**, 083701 (2012).
[2] Y. Ikeda, Y. Ito, S. Araki, T.C. Kobayashi, and H. Yoshizawa: J. Phys. Soc. Jpn. **84**, 024702 (2015).

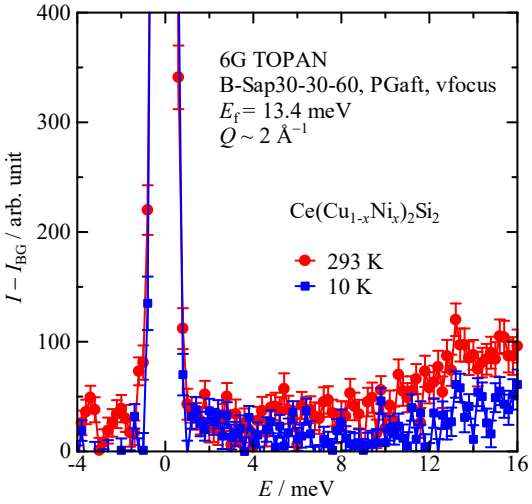

Fig. 1. Inelastic neutron scattering signal of $Ce(Cu_{1-x}Ni_x)_2Si_2$ measured at 10 and 293 K.